

REMARKS

Applicants have amended claim 1 to more particularly point out and distinctly claim the subject matter which they regard as their invention. No new matter has been introduced by the above amendment.

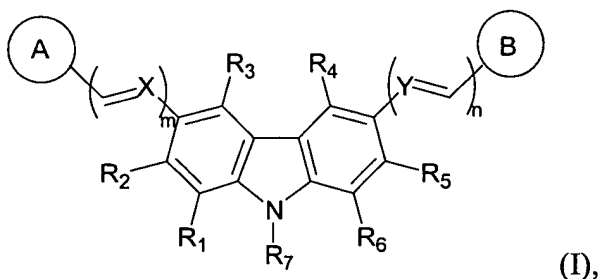
Claims 1-22 are currently pending. Reconsideration of the application, as amended, is requested in view of the remarks below.

The Examiner has allowed claims 12-22. See the Office Action, page 5, line 19. The Examiner has also concluded that claims 6-8 "would be allowable if rewritten in independent form including all of the limitations of the base claim and any intervening claims." See the Office Action, page 5, lines 15-18. On the other hand, the Examiner has rejected claims 1-5 and 9-11 under 35 U.S.C. § 102(b) or 35 U.S.C. § 103(a), relying on three prior art references. Applicants traverse each reference below.

The '864 patent

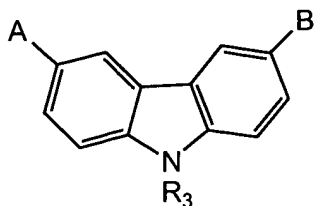
The Examiner rejects claims 1-3 and 9 as being anticipated by, or obvious over, Japanese Patent 2002-172864 ("the '864 patent"). See the Office Action, page 2, lines 11-13 and page 3, lines 14-16.

Amended claim 1 covers compounds of formula (I):



in which R₇ is H.

The '864 patent discloses carbazole compounds for use in an optical information recording medium. See the Abstract. Specifically, it discloses compounds having the following



formula: . In this formula, R₃ can be C1-C5 alkyl. The '864 patent does not disclose or suggest a carbazole compound in which R₃ is H. By contrast, R₇ in formula (I) of claim 1, corresponding to R₃ in the formula of the '864 patent, and can only be H, but not C1-C5 alkyl. In other words, as the '864 patent does not disclose the carbazole compounds covered by claim 1, it does not anticipate claim 1.

Further, the '864 patent does not even suggest the compounds of claim 1. Specifically, H is a non-alkyl group having a size much smaller than the C1-C5 alkyl groups. A carbazole compound in which R₇ is H would possess physical and chemical properties significantly different from those in which R₇ is C1-C5 alkyl. Thus, one skilled in the art would not have been motivated to replace C1-C5 alkyl (i.e., an alkyl group) with H (i.e., a non-alkyl group) to arrive at the carbazole compounds of claim 1. In other words, claim 1 is also not rendered obvious by the '864 patent.

In short, claim 1 is not anticipated or rendered obvious by the '864 patent. Neither are claims 2, 3, and 9, all of which depend from claim 1.

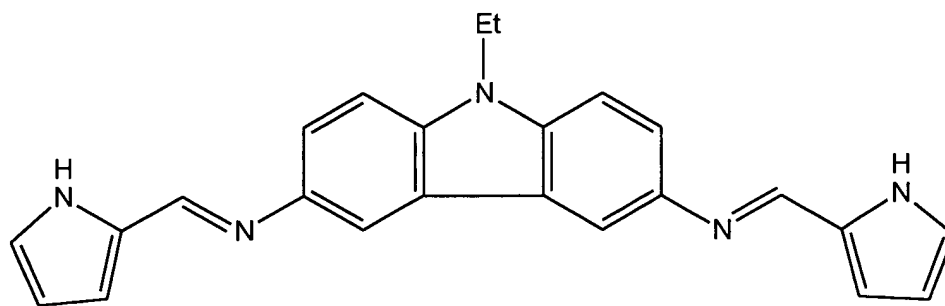
Even if a *prima facie* case of obviousness has been made (which Applicants do not concede), it can be successfully rebutted by a showing of an unexpected property of the carbazole compounds of claim 1. Specifically, the fluorescence intensity of the compounds of amended claim 1 increases significantly when binding to a DNA quadruplex. Example 9 of the Specification describes the fluorescence analysis of three exemplary compounds of claim 1 (i.e., compounds 2, 4, and 6) before and after binding to a DNA quadruplex. The results show that the fluorescence intensity of each compound unexpectedly increases by two orders of magnitude after binding to a DNA quadruplex. These compounds can be used as biomarkers for detecting DNA quadruplexes in electrophoresis. See page 11, line 23 to page 12, line 4. Given this unexpected property, claim 1 is clearly not obvious over the '864 patent on this additional ground. Neither are claims 2, 3, and 9, all of which depend from claim 1.

Krieg

The Examiner rejects claims 1-3 and 9 as being anticipated by, or obvious over, Krieg et al., Chemical Abstracts, 134:159233, 2000 ("Krieg"). See the Office Action, page 2, lines 11-13 and page 3, lines 14-16.

As mentioned above, amended claim 1 covers compounds of formula (I), in which R₇ is H.

Krieg discloses carbazole compounds that can be used as chromagens for detecting the activity of a peroxidase or related redox enzymes. See the Abstract. Specifically, the compound closest to those of amended claim 1 has the following formula:



In this compound, the group corresponding to R₇ in formula (I) of claim 1 is ethyl (i.e., an alkyl group). Similar to the '864 patent, Krieg does not disclose or even suggest a carbazole compound in which the group corresponding to R₇ is H (i.e., a non-alkyl group), as required by claim 1. Thus, claim 1 is not anticipated or rendered obvious by Krieg. Since claims 2, 3, and 9 depend from claim 1, they are also not anticipated or rendered obvious by Krieg.

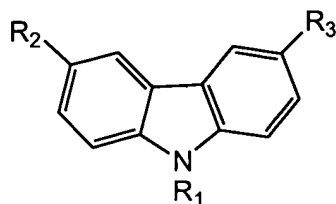
As discussed above, the Examiner's obviousness rejection can also be successfully rebutted by a showing of an unexpected property of the compounds of claim 1. Specifically, they exhibit a significant increase in fluorescence intensity after binding to a DNA quadruplex. Thus, claim 1, as well as claims 2, 3, and 9 dependent from it, is clearly not rendered obvious by Krieg on this additional ground.

Duan

The Examiner rejects claims 1-5 and 9-11 as being anticipated by, or obvious over, Duan et al., Chemical Abstracts, 134:107619, 2000 ("Duan"). See the Office Action, page 2, lines 20-22 and page 4, lines 15-17.

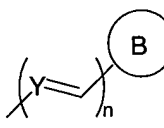
As mentioned above, amended claim 1 covers compounds of formula (I) in which R_7 is H. The Examiner relies on the abstract of Duan and asserts that “[t]he reference discloses the claimed compounds wherein ... R_7 is H ...” See the Office Action, page 2, line 23 to page 1, line 3. Applicants disagree. Applicants have obtained the full text of Duan, a copy of which is attached hereto as “Exhibit A.”

According to Exhibit A, Duan discloses carbazole compounds for use as novel ionic chromophores. The carbazole compounds have the following formula:



. See Figure 1 in Exhibit A. Referring to this formula, R_1 corresponds to R_7 in formula (I) of claim 1. Duan discloses that in two compounds that are the closest to those of amended claim 1, i.e., compounds 1a and 1b. See Figure 1. In these two compounds, R_1 is H.

Compound 1a is discussed first. R_3 in compound 1a is H. By contrast, the group in

formula (I) of claim 1 that corresponds to R_3 is . Thus, compound 1a differs substantially from the compounds of claim 1 and is therefore not covered by claim 1.

Compound 1b, on the other hand, is covered by amended claim 1. However, Duan does not provide an enabling disclosure of compound 1b. Specifically, Duan reports failure in the preparation of compound 1b: “For carbazole, diformylation was not proceeded.” See the paragraph bridging pages BB3.31.2 and BB3.31.3 of Exhibit A. According to MPEP § 2121.02, [w]here a process of making the compound is not developed until after the date of invention, the mere naming of a compound in a reference, without more, cannot constitute a description of the compound.” As Duan indicates that the preparation of compound 1b was unsuccessful, the mere naming of compound 1b does not constitute a description of this compound. It follows that compound 1b disclosed in Duan does not anticipate claim 1. Further, since there is a lack of known or obvious process of making compound 1b, this compound does not render claim 1 obvious. Applicants would like to point out that *In re Hoeksema*, 399 F.2d 269, 158 USPQ 597

(CCPA 1968) is a case in point. The *Hoeksema* court held that a prior art compound does not render a claimed compound obvious despite their structural similarity if there is a lack of known or obvious process to make the claimed compounds:

Thus, upon careful reconsideration it is our view that if the prior art of record fails to disclose or render obvious a method for making a claimed compound, at the time the invention was made, it may not be legally concluded that the compound itself is in the possession of the public. In this context, we say that **the absence of a known or obvious process for making the claimed compounds overcomes a presumption that the compounds are obvious**, based on close relationships between their structures and those of prior art compounds. Id. 601 (emphasis added).

Applicants prepared the compounds of claim 1 by reacting one equivalent of 3,6-dibromocarbazole with two equivalents of a heteroaryl compound containing a double bond (e.g., a 4-vinylpyridine) in the presence of a palladium catalyst. This method is not disclosed or suggested in Duan and therefore would not have been obvious over Duan. Thus, claim 1 is not rendered obvious by compound 1b in light of the *Hoeksema* holding.

In sum, as Duan does not disclose or suggest the compounds of claim 1, it does not anticipate or render obvious this claim. Since claims 2, 3, and 9 depend from claim 1, they are also not anticipated or rendered obvious by Duan.

As discussed above, the Examiner's obviousness rejection can also be successfully rebutted by a showing of an unexpected property of the compounds of amended claim 1. On this additional ground, claim 1, as well as claims 2, 3, and 9 dependent from it, is also not rendered obvious by Duan.

CONCLUSION

Applicants submit that the grounds for the rejection asserted by the Examiner have been overcome, and that claims 1-22, as pending, define subject matter that is novel and nonobvious over the prior art. On this basis, it is submitted that all claims are now in condition for allowance, an action of which is requested.

Applicant : Ta-Chau Chang et al.
Serial No. : 10/690,984
Filed : October 22, 2003
Page : 12 of 12

Attorney's Docket No.: 08919-112001 / 07A-920702

Please apply any other charges to deposit account 06-1050, referencing Attorney's
Docket No: 08919-112001.

Respectfully submitted,

Date: 12-17-04

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Linear and Nonlinear Optical Properties of Novel Ionic Chromophores

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ABSTRACT

A series of novel ionic chromophores consisting of a carbazole moiety and pyridinium rings connected by a double bond have been designed and synthesized as nonlinear optical materials. Their linear and nonlinear optical properties were investigated by semiempirical calculation and experiment. The absorption maximum wavelengths (λ_{max}) of these ionic chromophores showed shifting to longer wavelength region than their corresponding electrically neutral compounds. These ionic chromophores possess large first hyperpolarizabilities (β).

INTRODUCTION

π -Conjugated organic compounds have been widely studied as nonlinear optical (NLO) materials, which is expected to play a major role in various applications of NLO devices [1-2]. The optimization of NLO properties of organic materials is strongly related with both molecular and crystal engineering. In molecular level, stable molecules with large first hyperpolarizabilities (β) are requested to be designed and synthesized. These molecules should be aligned in optimized orientation so that the large β of a molecule is summed up to be large macroscopic second-order susceptibility, $\chi^{(2)}$. For second-order NLO materials, some guidelines of molecular design have been established. For example, those molecules with one π -conjugated system and donor and acceptor groups at the two side ends of the π -conjugated system (DA molecules) are useful to obtain large β values. Unfortunately, roughly 75% of all organic molecules crystallize in centrosymmetric space groups, leading to materials with vanishing $\chi^{(2)}$, e.g. second harmonic generation (SHG) [3]. To overcome this problem, using ionic π -conjugated chromophores could be useful to obtain noncentrosymmetric crystal structures [4]. It is because that ionic interactions could conquer dipole interactions, and numerous derivatives with different crystal structures could be prepared simply by changing the counter ions without any modification to the molecular structures of ionic π -conjugated chromophores. Along this line, many SHG active crystals of ionic chromophores including one series of stilbazolium cationic chromophores, i.e. 4-(2-(4-(dimethylamino)phenyl)ethenyl)pyridinium *p*-toluenesulfonate (DAST), have been found to exhibit significant second-order nonlinear optical susceptibilities ($\chi^{(2)}$) [4-8]. However, the β values of these cationic chromophores had not been evaluated until the hyper Rayleigh scattering (HRS) method was established [9]. In order to obtain the guideline of molecular design for ionic species with large β values, it was necessary to investigate the relationship between the molecular structures and β values. In our previous studies [10-12], the β values of some organic ions have been evaluated using HRS method and semiempirical quantum chemical calculation. It was figured out that stilbazolium cations have several time larger β values than their corresponding electrically neutral molecules. The large $\chi^{(2)}$ of

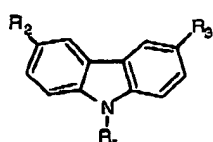
stilbazolium salts is due to the large β of stilbazolium cations. The significant electron withdrawing ability of pyridinium ring plays a major role in the intramolecular charge transfer of stilbazolium cations [12].

On the other hand, carbazole derivatives have been reported to have photoconductive, nonlinear optical and photorefractive properties [13-15]. Several types of crystals and polymers containing carbazole moieties have been obtained and showed reasonable second-order NLO properties [14-15]. Recently, we designed a new series of ionic chromophores, which was obtained by combining pyridinium ring and carbazole moiety using a double bond. In this paper, we report results on the linear and nonlinear optical properties of these novel ionic chromophores studied by semiempirical calculation and experimental measurement.

EXPERIMENTAL

The chemical structures of the ionic chromophores discussed in this paper (Compounds 1-3) were shown in Figure 1. In order to compare the properties of these ionic chromophores with those of their corresponding electrically neutral molecules, several electrically neutral carbazole derivatives (Compounds 4-5) also were discussed, which structures were shown in Figure 1. The semiempirical quantum chemical calculation with MNDO Hamiltonian, PM3 parameterization and Time-Dependence Hartree-Fock (TDHF) method were used to optimize the molecular geometries and evaluate the β values of these ionic chromophores and their corresponding electrically neutral molecules.

The synthesis schemes of several compounds were shown in Figure 2. All solvents and chemical reagents were used as received without further purification. 9*H*-Carbazole-3-carbaldehyde, 9-ethylcarbazole-3-carbaldehyde and 9-octylcarbazole-3,6-dicarbaldehyde were synthesized by a method previously reported [16-17]. These ionic chromophores, 1a, 2a and 3b, were synthesized simply by mixing these aldehyde and 1,4-dimethylpyridinium iodide in methanol under room temperature, respectively. For carbazole, diformylation was not proceeded, and for *N*-ethylcarbazole, solubility of dicarbaldehyde was too low though the compound was obtained. Thus, 1b and 2b have not been



Comp. No.	R ₁	R ₂	R ₃
1a	H	-CH=CH-Py ⁺ -CH ₃	-H
1b	H	-CH=CH-Py ⁺ -CH ₃	-CH=CH-Py ⁺ -CH ₃
2a	C ₂ H ₅	-CH=CH-Py ⁺ -CH ₃	H
2b	C ₂ H ₅	-CH=CH-Py ⁺ -CH ₃	-CH=CH-Py ⁺ -CH ₃
3b	C ₈ H ₁₇	-CH=CH-Py ⁺ -CH ₃	-CH=CH-Py ⁺ -CH ₃
4a	C ₂ H ₄ OH	-CH=CH-Ph-NO ₂	-H
4b	C ₂ H ₄ OH	-CH=CH-Ph-NO ₂	-CH=CH-Ph-NO ₂
5a	C ₂ H ₅	-NO ₂	-H
5b	C ₂ H ₅	-NO ₂	-NO ₂

Figure 1. Chemical structures of ionic chromophores 1-3 obtained by combining pyridinium ring and carbazole moiety through a double bond, and their corresponding electrically neutral molecules.

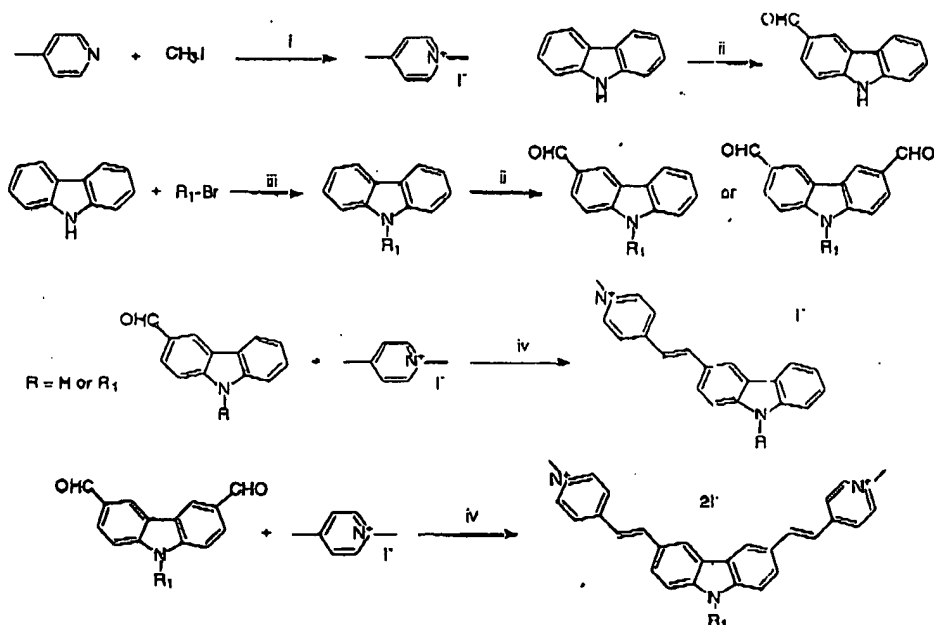


Figure 2. Synthesis schemes of ionic chromophores. i, MeOH, room temp., 12 h; ii, DMF-POCl₃, 90°C, 24 h; iii, NaOH, (PhCH₂)₂Et₃N⁺Cl, PhH, H₂O, reflux, 6 h; iv, MeOH, C₅H₅NH, room temp., 48 h.

prepared so far. The products were recrystallized several times from methanol. Structures of all compounds were confirmed by ¹H NMR and elemental analysis.

The UV/vis spectra of these ionic chromophores were obtained on a Shimadzu UV-3000 spectrometer from their methanol solutions of 10⁻⁵ mol/L. The fluorescence spectra were measured using a Shimadzu RF-5301PC spectrofluorometer.

RESULTS AND DISCUSSION

Carbazole derivatives are interesting organic π -conjugated molecules, which possesses both photoconductivity and second-order nonlinear optical response. The carbazole molecule has an isoelectronic structure with diphenylamine. Therefore, the introduction of electron-withdrawing groups in 3 and/or 6 positions induces intramolecular charge transfer (CT) and mesomeric dipole moment. The nitrogen atom of carbazole could be considered as a strong electron donor group. Because the pyridinium ring is a strong electron acceptor group, the combination of carbazole and pyridinium ring using a π -conjugated double bond should make an atypical DA molecule.

The UV/vis absorption spectra of ionic chromophores, **1a**, **2a** and **3b**, are shown in Figure 3. The maximum absorption wavelengths (λ_{max}) of the related compounds are summarized in Table I. The CT bands from 437 nm to 462 nm were observed for **1a**, **2a** and **3b**. The CT bands of these ionic chromophores were shifted to longer wavelength region than their corresponding electrically neutral molecules, **4a**, **5a** and **5b**. The disubstituted ionic carbazole **3b** showed an about 20 nm red-shifted λ_{max} , compared with the monosubstituted ionic species **2a**. The non-alkyl-substituted carbazole **1a** showed a

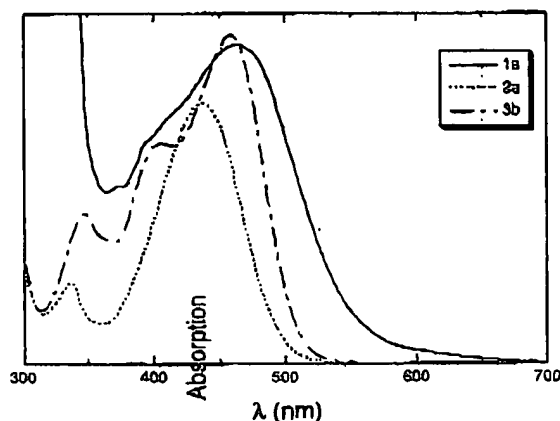


Figure 3. The UV/vis absorption spectra of ionic chromophores 1a, 2a and 3b, in methanol solution.

Table I. Data of the experimental maximum absorption wavelengths (λ_{max}), and calculated dipole moments in ground state (μ) and first hyperpolarizabilities (β_0).

Comp. No.	R ₁	R ₂	R ₃	λ_{max} (nm)	μ (D)	β_0 (10^{-30} esu)
DAST				474	12.73	241
1a	-H	-H	-CH=CH-Py ⁺ -CH ₃	462	16.74	228
1b	-H	-CH=CH-Py ⁺ -CH ₃	-CH=CH-Py ⁺ -CH ₃	—	14.22	181
2a	-C ₂ H ₅	-H	-CH=CH-Py ⁺ -CH ₃	437	17.42	253
2b	-C ₂ H ₅	-CH=CH-Py ⁺ -CH ₃	-CH=CH-Py ⁺ -CH ₃	—	16.51	197
3b	-C ₈ H ₁₇	-CH=CH-Py ⁺ -CH ₃	-CH=CH-Py ⁺ -CH ₃	458	—	—
4a	-C ₂ H ₄ OH	-H	-CH=CH-Ph-NO ₂	408 ^a	7.73	32
4b	-C ₂ H ₄ OH	-CH=CH-Ph-NO ₂	-CH=CH-Ph-NO ₂	—	10.99	38
5a	-C ₂ H ₅	-H	-NO ₂	374 ^a	7.08	6.0
5b	-C ₇ H ₁₅	-NO ₂	-NO ₂	370 ^a	8.92	5.5

^aData of λ_{max} were taken from ref. 14.

larger red-shift than alkyl-substituted ionic species 2a. For example, the λ_{max} of 1a and 2b were 462 nm and 437 nm, respectively. However, all the ionic species discussed here have shorter λ_{max} than DAST. Interestingly, a new CT band around 400 nm, which has not been observed for monosubstituted ionic species, was discovered only from the absorption spectrum of the disubstituted ionic chromophore (3b). This new CT band might be from the interaction between the two-subunits of the disubstituted ionic chromophores. All the ionic chromophores showed a fluorescence peak around 593 nm, when they

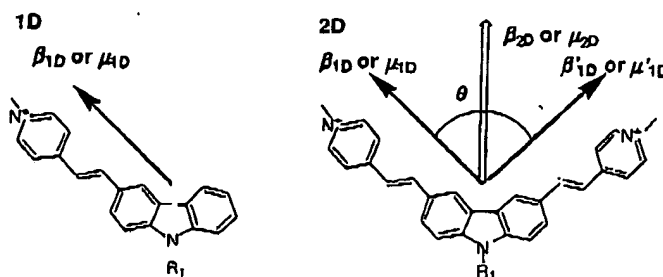


Figure 4. Model geometry of one-dimensional (1D) and two-dimensional (2D) charge transfer ionic π -conjugated molecules. In this figure, $|\mu_{1D}| = |\mu'_{1D}|$ and $|\beta_{1D}| = |\beta'_{1D}|$.

were excited at their λ_{max} .

The calculated dipole moments (μ) were summarized in Table I. Wada *et al.* reported the experimental μ values of several carbazole derivatives to be 6.4 D for 5a and 8.3 D for 5b, respectively [14]. Here, our calculated results, which showed that μ of 5a is 7.08 D and μ of 5b is 8.92 D, were in good agreement with the reported experimental values. Ionic chromophores 1 and 2 were calculated to have larger μ values than DAST, and they are about twice larger than their corresponding electrically neutral molecules. The monosubstituted carbazole derivatives such as 1a, 2a and 4a are classified into chromophores with one-dimensional charge transfer (1DCT), i.e. CT from carbazole ring to pyridinium ring. The disubstituted carbazole derivatives like 1b, 2b, 3b and 4b are said to be chromophores with dimensional charge transfer (2DCT), since the total charge transfer of the molecule is considered to be combination of two 1DCT of subunit from carbazole ring to two pyridinium ring in different directions. If we consider that the 2DCT is just simple vector summation of 1DCT, the μ values of 2DCT molecules (μ_{2D}) become larger than those of the corresponding 1DCT molecules (μ_{1D}). Because when the angle θ between two μ_{1D} s of the subunits are 120° , μ_{2D} equals to μ_{1D} , and θ was estimated to be less than 120° from the carbazole molecular structures. However, μ_{2D} values of 1b and 2b were less than μ_{1D} values of 1a and 2a, respectively. This indicated that, for these cases, electron donating ability of the carbazole ring is not sufficient to transfer electrons into two acceptors in the same rate as is the case of one acceptor. In the other words, electrons seem to be effectively transferred even for one pyridinium acceptor. On the other hand, μ_{2D} values of 4b and 5b were calculated to be more than μ_{1D} values of 4a and 5a, respectively. Namely, electron transfers from carbazole ring to nitro substituent do not so much occurs with that to pyridinium ring. This result is consistent with much larger μ_{1D} values of 1a and 2a than 4a and 5a.

The ionic chromophores showed several times larger β_0 values than their corresponding electrically neutral molecules. The monosubstituted compounds 1a and 2a had almost same β_0 values with DAST. The disubstituted ionic chromophores 1b and 2b always showed smaller β_0 values than the monosubstituted ionic chromophores 1a and 2a. Even for β_0 values, similar consideration to μ is possible. The β tensor component of 2DCT molecules (β_{2D}) should be larger than that of the corresponding 1DCT molecules (β_{1D}), if β_{2D} simple tensor summation of two β_{1D} . In the case of electrically neutral molecules, β_{2D} is more than β_{1D} for 4, and β_{2D} is near to β_{1D} for 5. However, β_{2D} values for 1b and 2b are less than 80% of β_{1D} values for 1a and 2a, respectively. From these results, disubstitution of pyridinium ring to carbazole ring is not so advantageous compared with its monosubstitution for increasing diagonal component of β , though diagonal component of $\chi^{(2)}$ may increase due to volume factor, i.e. 2DCT molecules should have the volume less than twice of that of

the corresponding IDCT molecules.

The investigations on crystal structures of these salts also have been done by changing their counter anions, and several crystals have been discovered to be active for second-order nonlinear optics. Detailed research work on single crystal growth and crystal structure analyses of these ionic chromophores is now in progress.

CONCLUSIONS

In conclusions, we designed a new series of ionic chromophores, and investigated their linear and nonlinear optical properties by semiempirical calculation and experiments. The novel ionic chromophores were designed by combining pyridinium rings and a carbazole moiety using a double bond. They have been discovered to possess strong absorption in visible region and large first hyperpolarizabilities. Because these novel ionic chromophores may possess both photoconductivity and large second-order nonlinear property, they could be expected to use as photorefractive and EO materials.

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